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Title of the Invention

A polyphenylene sulfide resin composition

What is claimed is:

1. A polyphenylene sulfide resin composition consisting of (B) 3 - 200 weight parts of one kind or two kinds or more selected from (B1) a polyolefin system polymer and/or a polyolefin system copolymer which satisfy the following equation 1, (B2) silicone and (B3) fluoro-resin, (C) 30-250 weight parts of magnesium hydroxide, (D) 0-350 weight parts of fibrous and/or non-fibrous fillers other than magnesium hydroxide, to (A) 100 weight parts of a polyphenylene sulfide resin.

[Equation 1]

$$S = \sum \theta_i ((\alpha_i + \beta_i + \gamma_i + \delta_i) / \omega_i) \geq 1.2$$

Wherein,  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ ,  $\delta_i$  and  $\omega_i$  are the number of atom or atomic group of the constitutional unit  $i$  in the polyolefin system copolymer, respectively, and  $\alpha_i$  represents the number of hydrogen atom,  $\beta_i$  the number of halogen atom,  $\gamma_i$  the number of carbonyl group,  $\delta_i$  the number of cyano group, and  $\omega_i$  the number of carbon atom, and further  $\theta_i$  represents a mol fraction of the constitutional unit  $i$  in the polyolefin system copolymer.  $\theta_i = 1$  in the case of homopolymer.

2. The polyphenylene sulfide resin composition according to claim 1, wherein the polyphenylene sulfide resin having the total ash content of 1.0 or less weight % is used as (A) the polyphenylene sulfide resin.
3. The polyphenylene sulfide resin composition according to claims 1 or 2, wherein the silicone is one or more kinds selected from silicone elastomer and silicone resin.
4. The polyphenylene sulfide resin composition according to any one of claims 1 to 3, wherein (E) organic silane compound are further loaded.
5. The polyphenylene sulfide resin composition according to claim 4, wherein the loadings of the organic silane compound are in the range of 0.1 - 5 weight parts, to (A) 100 weight parts of polyphenylene sulfide resin.
6. The polyphenylene sulfide resin composition according to claims 4 or 5, wherein the organic silane compound is one or two or more kinds selected from epoxy group content alkoxy silane compound, amino group content alkoxy silane compound, ureido

group content alkoxy silane compound, isocyanate group content alkoxy silane compound and mercapto group content alkoxy silane compound.

7. A molded article for electric appliance parts formed by injection-molding the polyphenylene sulfide resin composition according to any one of claims 1-6.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

This invention relates to a polyphenylene sulfide resin composition excellent in electrical properties, such as tracking resistance. It is especially applied to electrical-part such as electrical parts, electronic parts, or automobile electric parts, and also is applied to a wide range of various fields.

[0002]

[Description of the Prior Art]

A polyphenylene sulfide resin (it abbreviates to PPS resin below) has suitable properties such as excellent heat resistance, fire retardancy, rigidity, chemical resistance, electric insulation, and resistance to moist heat, as engineering plastics, and it is used in various kinds of electrical parts, electronic parts, machine parts, or automobile parts for, in particular, injection molding.

[0003]

However, the PPS resin has a defect that the tracking resistance is greatly inferior to other engineering plastics, such as a polyamide resin. Therefore, although the PPS resin has excellent properties as the electrical parts, such as the high heat resistance, the rigidity, the electric insulation, the fire retardancy, and the resistance to moist heat, the actual condition is that the application for a use which is exposed under comparatively high voltage is restricted.

[0004]

The trial of the tracking resistance improvement of the PPS resin has been heretofore made. For example, JP,64-69658,A discloses that the tracking resistance is improved by blending aliphatic series saturated ester consisting of long chain alcohol or short chain polyalcohol and long chain carboxylic acid, inorganic filler and reinforcing member with the PPS resin. However, the comparative tracking index (it abbreviates to CTI below) shown in the example is at most about 225V and it is difficult to say for the comparative tracking index to be fully improved.

[0005]

Although, on the other hand, JP,5-271542,A discloses that the tracking resistance is improved by blending polyamide and magnesium hydroxide with the PPS resin, the composition obtained has a large dimensional change at the time of water absorption, and its strength reduction under solution containing electrolytes, such as calcium chloride, is remarkable because of using a lot of polyamide. A tracking resistance property test is a test which evaluates the ease of happening of the dielectric breakdown under the harsh environment in which moisture and an electrolyte component exist, and therefore, the application of the composition to the use which requires high tracking resistance has a problem in view of long term reliability.

[0006]

[Problem to be solved by the Invention]

Then, this invention is to provide the PPS resin composition which is excellent in tracking resistance, without damaging various properties such as heat resistance, electric insulation, and low water absorption property which a PPS resin originally has.

[0007]

[Means for solving the Problem]

Namely, this invention is to provide a polyphenylene sulfide resin composition consisting of (B) 3 - 200 weight parts of one kind or two kinds or more selected from (B1) a polyolefin system polymer and/or a polyolefin system copolymer which satisfy the following equation 1, (B2) silicone and (B3) fluororesin, (C) 30-250 weight parts of magnesium hydroxide, (D) 0-350 weight parts of fibrous and/or non-fibrous fillers other than magnesium hydroxide, to (A) 100 weight parts of a polyphenylene sulfide resin, and a molded article for electric appliance parts formed by injection-molding the polyphenylene sulfide resin composition.

[0008]

[Equation 2]

$$S = \sum \theta_i ((\alpha_i + \beta_i + \gamma_i + \delta_i) / \omega_i) \geq 1.2$$

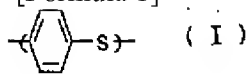
Wherein,  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ ,  $\delta_i$  and  $\omega_i$  are the number of atom or atomic group of the constitutional unit  $i$  in the polyolefin system copolymer, respectively, and  $\alpha_i$  represents the number of hydrogen atom,  $\beta_i$  the number of halogen atom,  $\gamma_i$  the number of carbonyl group,  $\delta_i$  the number of cyano group, and  $\omega_i$  the number of carbon atom, and further  $\theta_i$  represents a mol fraction of the constitutional unit  $i$  in the polyolefin system copolymer.  $\theta_i = 1$  in the case of homopolymer.

[0009]

[The mode of implementation of the invention]

The polyphenylene sulfide resin used in this invention is a polymer comprising more than 70 mol %, preferably more than 90 mol %, of repeating unit represented by the structural-formula [I]

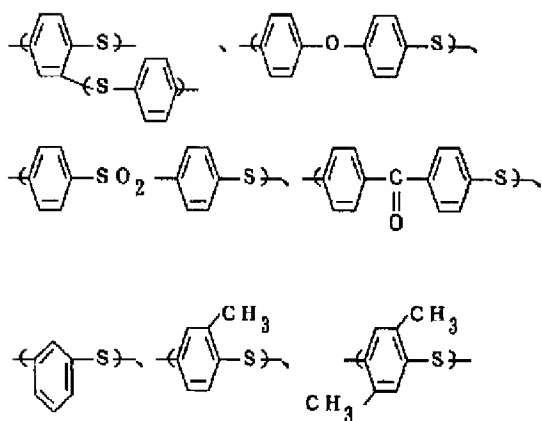
[Formula 1]



The polymer comprising the above repeating unit in an amount smaller than 70 mol % is not preferred because the heat resistance is lowered. Further, the PPS resin can comprise up to 30 mol % of repeating units represented by one or more of the following structural formulae;

[0010]

[Formula 2]



As to the melt viscosity of the PPS resin used in this invention, there will be no restriction in particular, if it is possible to melt- knead. However, in general, the PPS resin having the melt viscosity of in the range of 50-20,000 poise (320 °C, shear rate of 1000 sec<sup>-1</sup>) is used, preferably in the range of 100-2000 poise and most preferably in the range of 200-1000 poise.

[0011]

This PPS resin can be manufactured by the method of obtaining the polymer with a relatively high molecular weight described in JP,52-12240,B or JP,61-7332,A, or the method of obtaining the polymer with a relatively low molecular weight described in JP,45-3368,B, and these methods are known. The PPS resin obtained as mentioned above in this invention can be also used, after applying various processes such as crosslinking/high-polymerization by heating in the air, heat treatment under the inert gas atmosphere of nitrogen etc. or decompression, washing by organic solvent, hot water, acid aqueous solution, etc., activation by compounds containing functional group acid anhydride, amine, isocyanate and disulfide compound containing functional group.

[0012]

Further, in the PPS resin composition of this invention, to use the PPS resin having the amount of total ash content of 1.0 weight % or less, preferably 0.8 weight % or less, most preferably 0.5 weight % or less, is suitable in view of obtaining the resin composition having much excellent tracking resistance. Although the reason for this is not clear, it is assumed that the elution of an ionic substance in the PPS resin measured as the amount of total ash content affect adversely to the tracking resistance. The amount of total ash content is the one that the ratio of the residue weight to dried resin (5g) is calculated by putting resin 5g dried at 150 °C for 1 hour into a crucible and measuring the residue weight of the resin burned for 6 hours at 540 °C.

[0013]

An ingredient (B1) used in this invention is a polyolefin system polymer and/or a polyolefin system copolymer which satisfy the following expression.

[0014]

[Equation 3]

$$S = \sum \theta_i ((\alpha_i + \beta_i + \gamma_i + \delta_i) / \omega_i) \geq 1.2$$

Wherein,  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ ,  $\delta_i$  and  $\omega_i$  are the number of atom or atomic group of the constitutional unit  $i$  in the polyolefin system copolymer, respectively, and  $\alpha_i$  represents the number of hydrogen atom,  $\beta_i$  the number of halogen atom,  $\gamma_i$  the number of carbonyl group,  $\delta_i$  the number of cyano group, and  $\omega_i$  the number of carbon atom, and further  $\theta_i$  represents a mol fraction of the constitutional unit  $i$  in the polyolefin system copolymer.  $\theta_i = 1$  in the case of homopolymer.

As an example of this (B1) polyolefin system polymer and/or a polyolefin system copolymer, we can show homopolymer obtained by polymerization of unsaturated monomer such as alpha olefins containing 2-20 carbon atoms such as ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 3-methyl-1-butene, 3,3-dimethyl-1-butene, 3-methyl-1-pentene, 4-methyl-1-pentene, 4,4-dimethyl-1-pentene, 3-methyl-1-hexene, 4-methyl-1-hexene, 5-methyl-1-hexene, 1-octene, 1-decene, allyl cyclopentane, allylcyclohexane, 3-cyclohexyl-1-butene, vinylcyclopropane, vinylcyclohexane, and 2-vinylbicyclo[2,2,1]-heptane, nonconjugated diene compounds such as dicyclopentadiene, 1,4-hexadiene, cyclo-octadiene and methylene norbornene, conjugated diene compounds such as butadiene and isoprene, vinylic halide compounds such as PVC and vinylidene chloride and the random copolymer, block copolymer and graft copolymer. Further, the above-mentioned unsaturated monomer and unsaturated monomers other than the above, for example, unsaturation organic acid and its derivatives such as acrylic acid, methacrylic acid, etah kurile acid, itaconic acid, maleic acid, methyl acrylate, methyl methacrylate, ethyl acrylate, ethyl methacrylate, etah kurile acid methyl, glycidyl acrylate, glycidyl methacrylate, glycidyl ethacrylate, maleic acid anhydride, acrylic acid metal salt, methacrylic acid metal salt, etah kurile acid metal salt and itaconic acid metal salt, vinyl ester such as vinyl acetate and butanoic acid vinyl, vinylsilane such as vinyl trimethylmethoxysilane and gamma-methacryloyl oxypropyl trimethoxysilane, vinyl ether such as vinylmethyl ether, vinylethyl ether and vinyl isobutyl ether, aromatic vinyl compounds such as styrene, etc., it is also available to apply random copolymer, block polymer and graft copolymer with the above. As a concrete example of these (B1) polyolefin system polymers and/or polyolefin system copolymer, high density polyethylene, low density polyethylene, straight-chain-shape low density polyethylene, polypropylene, ethylene/propylene copolymer, ethylene / propylene / butadiene copolymer, ethylene/vinyl acetate copolymer, ethylene/ethyl acrylate copolymer, ethylene / ethyl methacrylate copolymer, ethylene / sodium methacrylate copolymer, ethylene / methacrylic-acid-zinc copolymer, styrene/butadiene copolymer, styrene / butadiene / styrene copolymer, styrene / ethylene / butylene / styrene copolymer, ethylene / glycidyl acrylate copolymer, ethylene / glycidyl methacrylate copolymer, acrylonitrile/styrene copolymer which is carried out the graft copolymerization of ethylene/glycidyl methacrylate, the polystyrene polymer which is carried out the graft copolymerization of ethylene/glycidyl methacrylate, the polymethylmethacrylate polymer etc. which is carried out the graft copolymerization of ethylene/glycidyl methacrylate are shown, and especially high density polyethylene, low density polyethylene, straight-chain-shape low density polyethylene, ethylene / propylene copolymer, ethylene/ethyl acrylate copolymer, ethylene / glycidyl methacrylate copolymer, acrylonitrile/styrene copolymer that is carried out the graft copolymerization of ethylene/glycidyl methacrylate, and ethylene / ethyl methacrylate

copolymer are preferred.

[0015]

These (B1) polyolefin system polymers and/or polyolefin system copolymer can be used together in two or more kinds of polymers, and also (B3) can be used together with fluororesin and/or (B2) silicone.

[0016]

Loadings of this (B1) polyolefin system polymer and/or polyolefin system copolymer is in the range of 3 to 200 weight parts, preferably in the range of 5 to 100 weight parts, most preferably in the range of 10 to 70 weight parts, to 100 weight parts of the PPS resin. If the loadings is less than 3 weight parts, the improvement of the tracking resistance is insufficient and is not preferable. If the loadings is more than 200 weight parts, the mechanical strength of the resin composition is damaged and therefore is not preferable.

[0017]

The polyolefin system polymer and/or the polyolefin system copolymer used in this invention (B1) need to satisfy the following expression.

[Equation 4]

$$S = \sum \theta_i ((\alpha_i + \beta_i + \gamma_i + \delta_i) / \omega_i) \geq 1.2$$

Wherein,  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ ,  $\delta_i$  and  $\omega_i$  are the number of atom or atomic group of the constitutional unit  $i$  in the polyolefin system copolymer, respectively, and  $\alpha_i$  represents the number of hydrogen atom,  $\beta_i$  the number of halogen atom,  $\gamma_i$  the number of carbonyl group,  $\delta_i$  the number of cyano group, and  $\omega_i$  the number of carbon atom, and further  $\theta_i$  represents a mol fraction of the constitutional unit  $i$  in the polyolefin system copolymer.  $\theta_i = 1$  in the case of homopolymer.

Even if the polyolefin system copolymer not satisfying the expression is used as the ingredient (B1) of this invention, the satisfactory tracking resistance is not obtained. As a polyolefin system copolymer unsuitable for this invention, for example, homopolymer of styrene and methylstyrene, or copolymer with high content of styrene, methylstyrene and maleic anhydride, etc. are shown. In the above-mentioned expression, it is  $S \geq 1.2$ , however,  $S \geq 1.5$  is more preferred. Wherein, explaining the above-mentioned expression, in the case of ethylene unit, since the number of hydrogen atoms is 4 and the number of carbon atoms is 2, it is set to  $S = 2.00$ . In the case of acrylic acid unit, since the number of hydrogen atoms is 4, the number of carbonyl groups is 1 and the number of carbon atoms is 3, it is set to  $S = 1.67$ . Further, for example, in the case of ethylene / acrylic acid (mole ratios 7/3) copolymer, it is set to  $S = 2.00 \times 0.7 + 1.67 \times 0.3 = 1.90$ .

[0018]

(B2) silicone used in this invention is the organic silicon compound which has the siloxane bond as a skeleton and in which the organic group etc. is directly coupled with its silicon. As the organic group coupled directly with silicon, although methyl group, ethyl group, phenyl group, vinyl group, trifluoropropyl group and those concomitant use are known, these known silicone can be especially used without restriction.

Silicone in which a part of organic group was replaced by a substituent having epoxy group, amino group, polyether group, carboxyl group, mercaptogroup, ester group, chloro alkyl group, alkyl group of three or more carbon numbers, hydroxyl, etc. can be also used. Silicone is classified into silicone oil, silicone elastomer and silicone resin from the degree of the crosslinking. (As to silicone, it is described in detail in "silicone material handbook" (the Dow Corning Toray Silicone Inc. issued and edited, August, 1993). Although any kinds of silicone can be used as the silicone of this invention, the silicone elastomer and the silicone resin are preferable from a viewpoint of the mechanical strength, the workability and the tracking resistance, etc. Silicone elastomer and silicone resin (for example, "Trefil "E, R series manufactured by Dow Corning Toray Silicone Inc.) with a mean particle diameter of 0.1-80  $\mu\text{m}$  which were finely powdered is available in view of good dispersibility and excellent workability. Further, these (B-2) silicone can be also used together in two or more kinds. Concomitant use with a polyolefin (B1) system copolymer and/or (B3) fluoro resin is also possible.

[0019]

Loadings of this (B2) silicone is in the range of 3 to 200 weight parts, preferably in the range of 5 to 100 weight parts, most preferably in the range of 10 to 70 weight parts, to 100 weight parts of the PPS resin. If the loadings is less than 3 weight parts, the improvement of the tracking resistance is insufficient and is not preferable. If the loadings is more than 200 weight parts, the mechanical strength of the resin composition is damaged and therefore is not preferable.

[0020]

(B3) Fluoro resin used in this invention is a resin which contains a fluorine atom in the resin. As the example, polytetrafluoroethylene, polychlorotrifluoroethylene, polyvinylidene fluoride, polyvinyl fluoride, tetrafluoroethylene / hexafluoropropylene copolymer, tetrafluoroethylene / perfluoroalkyl vinyl ether copolymer, tetrafluoroethylene/ethylene copolymer, tetrafluoroethylene / hexafluoropropylene / perfluoroalkyl vinyl ether copolymer, tetrafluoroethylene / hexafluoropropylene / vinylidene fluoride copolymer, chlorotrifluoroethylene/ethylene copolymer, chlorotrifluoroethylene/vinylidene fluoride copolymer, vinylidene fluoride / hexafluoropropylene copolymer, fluorinated ethylene/propylene copolymer, etc. can be shown. A copolymerizable monomer together with the above-mentioned fluoro resin, to such an extent that the effect of this invention is not damaged, and a copolymer obtained by polymerizing in using together with end sealing agent can be also available. Further, these (B3) fluoro resin can be used together in two or more kinds. Concomitant use with a polyolefin system (B1) copolymer and/or (B2) silicone is also possible.

[0021]

Loadings of this (B3) fluoro resin is in the range of 3 to 200 weight parts, preferably in the range of 5 to 100 weight parts, most preferably in the range of 10 to 70 weight parts, to 100 weight parts of the PPS resin. If the loadings is less than 3 weight parts, the improvement of the tracking resistance is insufficient and is not preferable. If the loadings is more than 200 weight parts, the mechanical strength and the fluidness etc. of the resin composition is damaged and therefore is not preferable.

[0022]

When using together two or more sorts of (B1) - (B3), the total amount of the ingredient of (B1) - (B3) is loaded in the range of 3 to 200 weight parts.

[0023]

As (C) magnesium hydroxide used in this invention, Magnesium hydroxide with relatively high purity which contains an inorganic substance more than 80 weight % shown by chemical formula  $\text{Mg}(\text{OH})_2$ . A magnesium hydroxide with high purity is suitable from a viewpoint of the tracking resistance, the mechanical strength and the melt viscosity, which preferably contains the inorganic substance more than 80 weight % shown by  $\text{Mg}(\text{OH})_2$ , CaO less than 5 weight % and chlorine less than 1 weight %, and further which more preferably contains  $\text{Mg}(\text{OH})_2$  more than 95 weight %, CaO less than 1 weight % and chlorine less than 0.5 weight %, and furthermore which most preferably contains  $\text{Mg}(\text{OH})_2$  more than 98 weight %, CaO less than 0.1 weight % and chlorine less than 0.1 weight %.

[0024]

The shape of magnesium hydroxide used in this invention may be in a particle state, a flake state and a fibrous state. The particle state and the flake state are most preferred from a viewpoint of the dispersibility etc. Further, it is preferred that the specific surface area is less than  $15\text{-m}^2/\text{g}$ , preferably less than  $10\text{-m}^2/\text{g}$ . If the specific surface area is more than  $15\text{-m}^2/\text{g}$ , the improvement of the tracking resistance and the mechanical strength etc. are damaged and it is not preferable, the reason for this is probably that it will influence the dispersibility of magnesium hydroxide. In the case of particle state and the shape of flakes, the mean particle diameter in the range of  $0.3\text{-}10\text{ }\mu\text{m}$ , preferably in the range of  $0.3\text{-}2\text{ }\mu\text{m}$  is preferable from a viewpoint of the tracking resistance, the mechanical strength and the melt viscosity. Further, in the case of using the fibrous state, the mean fiber diameter of  $0.1\text{-}2\text{ }\mu\text{m}$  and the aspect ratio of 20-60 is suitable, and especially the mean fiber diameter of  $0.3\text{-}2\text{ }\mu\text{m}$  and the aspect ratio of 30-50 is preferably suitable.

[0025]

It is preferred that this (C) magnesium hydroxide is surface-treated to use by vinylsilane compounds such as vinyltriethoxysilane and vinyl trichlorosilane, epoxysilane compounds such as gamma-glycidoxypropyltrimethoxysilane, gamma-glycidoxy propyltriethoxysilane and beta-(3, 4-epoxycyclohexyl) ethyltrimethoxysilane, amino silane compounds such as gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, gamma-(2-aminoethyl) aminopropyl trimethoxysilane and gamma-aminopropyl trimethoxysilane and long chain fatty acid or long chain fatty alcohol such as stearic acid, oleic acid, montanic acid and stearyl alcohol. Especially application of (C)magnesium hydroxide that is surface-treated with epoxysilane compound and amino silane compound is suitable in respect of the improvement of the tracking resistance and the mechanical strength.

[0026]

Loadings of this (C) magnesium hydroxide is in the range of 30 to 250 weight parts, preferably in the range of 30 to 150 weight parts, most preferably in the range of 50 to 150 weight parts, to 100 weight parts of the PPS resin. If the loadings is less than 30 weight parts, the improvement of the tracking resistance is insufficient and is not preferable. If the loadings is more than 250 weight parts, the mechanical strength and the fluidness etc. of the resin composition is damaged and therefore is not preferable.

[0027]

In this invention, in order to improve the strength and the dimensional stability, etc.,



the fibrous filler and/or the non-fibrous filler other than (D) magnesium hydroxide is used in response to the necessity. As the fibrous filler and/or the non-fibrous filler other than this (D) magnesium hydroxide, fibrous fillers such as glass fiber, carbon fiber, titanate potassium whisker, zinc oxide whisker, boric acid aluminum whisker, aramid fiber, alumina fiber, silicon carbide fiber, ceramic fiber, asbestos fiber, stone coe textiles, and metal fiber, silicate such as zeolite, sericite, kaolin, mica, clay, pyrophyllite, bentonite, asbestos, talc, and alumina silicate, metallic compounds such as alumina, oxidized silicon, magnesium oxide, zirconium oxide, titanium oxide, iron oxide, carbonate such as calcium carbonate, magnesium carbonate and dolomite, sulfate such as calcium sulfate and barium sulfate, non-fibrous fillers such as glass bead, ceramic bead, boron nitride, silicon carbide and silica may be shown. These may be hollow and may be used together in two or more kinds of the fillers. Further, It is preferred that the fibrous fillers and/or the non-fibrous filler other than these (D) magnesium hydroxide is pre-treated to use by coupling agents such as isocyanate system compound, the organic silane system compound, organic titanate compound, organic borane system compound, epoxy compound in order to obtains excellent mechanical strength.

[0028]

The loadings of the fibrous fillers and/or the non-fibrous fillers other than this (D) magnesium hydroxide is selected in the range of 0 - 350 weight parts to 100 weight parts of the (A)PPS resin. Further, the loadings of the fibrous fillers and/or the non-fibrous fillers is preferably selected in the range of 20 - 200 weight parts to 100 weight parts of the (A)PPS resin in order to obtain the resin composition which is more excellent in a balance of the melting fluidness and the mechanical strength.

[0029]

The loadings of the organic silane compound (E) to the PPS resin composition of this invention is effective in restricting the erosion at the time of tracking represented by restricting the erosion at the time of a tracking test, and also effective in improving the mechanical strength and the moldability, etc. As the example of this organic silane compound, epoxy group content alkoxy silane compounds such as gamma-glycidoxypolytrimethoxysilane, gamma-glycidoxypolytriethoxysilane and beta-(3,4-epoxycyclohexyl) ethyltrimethoxysilane, mercapto group content alkoxy silane compounds such as gamma-mercapto polytrimethoxysilane, gamma-mercaptopolytriethoxysilane, ureido group content alkoxy silane compounds such as gamma-ureido polytriethoxysilane, gamma-ureido polytrimethoxy silane, gamma-(2-ureido ethyl) aminopropyl trimethoxysilane, isocyanato group content alkoxy silane compounds such as gamma-isocyanatopropyl triethoxysilane, gamma-isocyanato polytrimethoxysilane, gamma-isocyanato polydimethyl dimethoxysilane, gamma-isocyanato polydimethyl diethoxysilane, gamma-isocyanatopropylethyl dimethoxysilane, gamma-isocyanatopropylethyl diethoxysilane and gamma-isocyanatopropyltrichlorosilane, amino group content alkoxy silane compounds such as gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, gamma-(2-aminoethyl) aminopropyl trimethoxysilane and gamma-aminopropyl trimethoxysilane can be shown. The loadings of this organic silane compound is preferably in the range of 0.1 - 5 weight parts to 100 weight parts of (A) PPS resin, and most preferably in the range of 0.2 - 2 weight parts.

[0030]

In the range of not damaging the effect of this invention, to the PPS resin composition of this invention, plasticizers such as polyalkylene oxide oligomer system compound,

thioether series compound, ester system compound and organophosphorus compound, nucleus agents such as talc, kaolin and organophosphorus compound, release agents such as polyolefin system compound, silicone series compound, and long chain aliphatic series ester system compound, antioxidant, thermostabilizer, lubricant, ultraviolet inhibitor, colorant, fire retardant, and foaming agent can be added.

The PPS resin composition of this invention can contain the resin such as polyester, polyphenylene oxide, polysulfone, polyether imide, polyamidoimide, polyimide, polycarbonate, polyether sulphone, polyether ketone, polythioether ketone, polyether ether ketone, epoxy resin, phenol resin, polyamide elastomer, polyester elastomer and polyalkylene oxide, in the range of not damaging the effect of this invention, However, as described in the prior art, concomitant use of polyamide resin must be avoided.

Especially, concomitant use of a lot of polyamide resin of 10 or more weight parts to 100 weight parts of the PPS resin as shown in JP,5-271542,A must be avoided.

Therefore, concomitant use of polyamide resin is less than 10 weight parts to 100 weight parts of the PPS resin and usually about 5 or less weight parts, preferably not contain substantially the polyamide resin.

[0031]

Although there is no restriction of a method of preparing the PPS resin composition of this invention, as an example, a method of supplying the mixture of raw material to the melting mixers such as a single screw or a double screw extruder, a Banbury mixer, a kneader and a roll mill, these being publicly known, and kneading at temperature of 280-380 °C can be shown. There is also no restriction of mixed order of the raw material, for example, there is a method of melt kneading by the methods as mentioned above after the loadings of all the raw materials, a method of melt kneading by the methods as mentioned above after the loadings of a part of the raw materials and the loadings of the remaining raw materials. It is also possible to add the small-quantity additive component before molding after other ingredients are kneaded and pelletized by the method mentioned above.

[0032]

The PPS resin composition obtained by this invention is the resin composition having the new property of improved tracking resistance, which was insufficient for the conventional PPS resin, without damaging thermal stability, melting fluidity, mechanical strength, electric insulation, and low water absorption property which the PPS resin composition originally own. The PPS resin composition thus obtained can be applied to publicly known molding methods such as injection molding, extrusion molding, compression molding, blow forming, injection compression molding, etc. Especially, it is a suitable resin composition for injection molding. The molded article of the PPS resin composition of this invention is suitable for a various applications such as electric appliance part of a dynamo, an electric motor, a transformer, a current transmission, a voltage regulator, a rectifier, an inverter, a relay, a point of contact for electric power, a switch, a breaker, a knife-switch, other pole rods, an electrical-part cabinet, a light socket, a plug, and a power module as well as Electronic parts such as a sensor, a LED lamp, a connector, a resistor, a relay case, a miniature switch, a coil bobbin, a capacitor, a variable condenser case, an optical pickup, an oscillator, a transformer, a printed circuit board, a tuner, a speaker, a microphone, a headphone, a miniature size motor, a magnetic head base, a semiconductor, a liquid crystal, a FDD carriage, a FDD chassis, a motor brush electrode holder, a parabolic antenna, computer

associated parts etc.; audio apparatus part such as a VTR part article, a television part, an iron, a hair drier, a rice cooker part, a microwave oven part, an audio component, an audio laser disc compact disk, a home, clerical work electric product parts such as a lighting part, a refrigerator part, an air-conditioner part, a typewriter part, a word processor part; a machinery associated part such as an office computer associated part, a telephone associated part, a facsimile associated part, a copying machine associated part, a jig for washing, a motor part article, a writer, a typewriter: an optical instrument, precision instrument associated parts such as a microscope, binoculars, a camera, a clock; a alternator terminal, a alternator connector, an I.C. regulator, a potentiometer base, various valves such as an exhaust gas valve, an exhaust system and a suction system pipe, an air intake nozzle snorkel, an intake manifold, a fuel pump, an engine-cooling-water joint, a carburetor main body, a carburetor spacer, an exhaust gas sensor, a cooling water sensor, an oil temperature sensor, a brake pad wear sensor, a throttle position sensor, a crankshaft position sensor, an air flow meter, a brake-friction-pad wear sensor, a thermostat base for air-conditioners, a heating warm air flow controlling valve, a brush electrode holder for radiator motors, a water pump impeller, a turbine vein, a windshield-wiper-motor relation part, a distributor, a starting switch, a starter relay, a wire harness for transmission, a window washer nozzle, an air conditioning panel switch board, a coil for fuel relation electromagnetism valves, a connector for fuses, a horn terminal, an electric insulating plate of electric equipment article, a stepping motor rotor, a lamp socket, a lamp reflector, a lamp housing, a brake piston, a solenoid bobbin, an engine oil filter, and an ignition case, etc.

[0033]

[Example]

An example is given to below and this invention will be explained in detail.

[0034]

The tracking resistance and the bending strength were measured in accordance with the following methods, which were shown in the example and the comparative example.

[0035]

The tracking test: The composition is dry-blended and this was pelletized with the 40 mm single screw extruder. The ASTM No. 1 specimen was injection-molded from the obtained pellet by the use of the injection molding machine with a maximum clamping pressure power of 75 t. In accordance with the IEC112 method, the comparative tracking index (CTI) was measured using this specimen.

[0036]

Bending strength: it was in accordance with ASTM D790.

[0037]

Reference example 1 (PPS resin)

In the example and the comparative example, M2888 (PPS-1) and M2900 (PPS-2) manufactured by Toray Industries, Inc., were used as the PPS resin. The amount of total ash of this PPS resin was 0.07 weight % and 0.68 weight %, respectively.

[0038]

Reference example 2 (polymerization of PPS)

4.67 kg of 30% hydrosulfide solution, 2.00 kg of 50% sodium hydroxide, and NMP8kg are put into an autoclave, this is gradually raised up to a temperature of 205 °C with agitation, and the distillate water 4.1l containing 3.8 kg of water was removed. 3.75 kg of 1,4-dichlorobenzene and NMP2kg were added to the remains mixture, and it heated

at 230 °C for 1 hour and further at 260 °C for 2 hours. The obtained product was filtered, rinsed 3 times at the room temperature and dried at 80 °C for 24 hours under reduced pressure condition, and thus the PPS resin (PPS-3) of the powder of the number average molecular weight 5400 was obtained. The amount of total ash of this PPS resin was 1.20 weight %.

[0039]

Examples 1-4

After (A) PPS resin, (B) polyolefin system copolymer, copolymer selected from silicone and fluororesin, (C) magnesium hydroxide, and (D) filler are dry blended at a rate shown in Table 1, it is pelletized after melt kneading with the single screw extruder set to the temperature conditions 280-320 °C. The test piece for measurement of the tracking resistance and the bending strength was fabricated, using the obtained pellet. The CTI and the bending strength measured in relation to the obtained test piece are shown in Table 1.

[0040]

Examples 5-9

After (A) PPS resin (PPS-1 - 3), (B) polyolefine system polymer, (C) magnesium hydroxide, and (D) filler, (E) organic silane compound are dry blended at a rate shown in Table 1, it is pelletized after melt kneading with the single screw extruder set to the temperature conditions 280-320 °C. The test piece for measurement of the tracking resistance and the bending strength was fabricated, using the obtained pellet. The CTI and the bending strength measured in relation to the obtained test piece are shown in Table 1.

[0041]

Even if any PPS resin of PPS-1 - PPS-3 was used, the improvement in the tracking resistance can be obtained, however, the effect appears more notably by the use of the PPS resin having less amount of total ash.

[0042]

Comparative example 1 – 3

The dry blend, the melt kneading and the pelletizing were performed like Examples 1-7 except not adding the resin selected from (B) polyolefin system copolymer, silicone and fluororesin. The test piece for measurement of the tracking resistance and the bending strength was fabricated, using the obtained pellet. The CTI and the bending strength measured in relation to the obtained test piece are shown in Table 2.

[0043]

Thus, the CTI was greatly improved by blending the resin selected from (B) polyolefin system copolymer, silicone and fluororesin.

[0044]

Comparative example 4

The dry blend, the melt kneading and the pelletizing were performed like Examples 1-3 except using polystyrene (S= 1.0) as (B) ingredients. The test piece for measurement of the tracking resistance and the bending strength was fabricated, using the obtained pellet. The CTI and the bending strength measured in relation to the obtained test piece are shown in Table 2.

[0045]

Even if the polyolefine system polymer having a low S value of 1.0 like polystyrene are used, the improvement of sufficient tracking resistance can not be obtained.

[0046]

[Table 1]

[0047]  
[Table 2]

	Example 1	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7
Kind of PPS(A) Loadings (weight part)	PPS-1 25	PPS-1 25	PPS-1 25	PPS-1 27	PPS-1 30	PPS-2 30	PPS-3 30
Kind of resin(B) Loadings (weight part)	B-1 15	B-2 15	B-3 15	B-1 5	B-1 10	B-1 10	B-1 10
Kind of magnesium hydroxide(C) Loadings (weight part)	C-1 30	C-1 30	C-1 30	C-1 24	C-1 35	C-1 35	C-1 35
Kind of filler(D) Loadings (weight part)	glass fiber 30	glass fiber 30	glass fiber 30	glass fiber/talc 40/4	glass fiber 25	glass fiber 25	glass fiber 25
Kind of organic silane compound(E) Loadings (weight part)	---	---	---	---	---	---	---
CTI	800	600	600	550	600	550	475
Bending strength (kgf/cm <sup>2</sup> )	1410	1380	1500	1580	1320	1270	1110

B-1: Polyethylene (manufactured by Mitsui Petro-Chemical Inc. "highzecs" 1300J)

B-2: "Trefil" R902 manufactured by Dow Corning Toray Silicone Inc.

B-3: Polytetrafluoroethylene (manufactured by Daikin Industry Inc. "Lubron" L-5)

C-1: Magnesium hydroxide (Mg(OH)<sub>2</sub> 98.8wt%, CaO 0.05 wt%, Cl 0.04wt%)

E-1: beta-(3,4-epoxycyclohexyl) ethyltrimethoxysilane

E-2: gamma-(3,4-epoxycyclohexyl) ethyltrimethoxysilane

	Compar at i ve Exampl e 1	Compar at i ve Exampl e 2	Compar at i ve Exampl e 3	Compar at i ve Exampl e 4
Ki nd of PPS(A) Loadi ngs ( wei ght part )	P P S - 1 4 0	P P S - 1 3 2	P P S - 1 4 0	P P S - 1 2 5
Ki nd of resi n(B) Loadi ngs ( wei ght part )	----	----	----	B - 4 1 5
Ki nd of magnesi um hydroxi de(C) Loadi ngs ( wei ght part )	C - 1 3 0	C - 1 2 4	C - 1 3 5	C - 1 3 0
Ki nd of filler(D) Loadi ngs ( wei ght part )	gl ass fi ber 30	gl ass fi ber / tal c 40/ 4	gl ass fi ber 25	gl ass fi ber 30
C T I	275	275	250	375
Bendi ng strengt h ( kgf / cm <sup>2</sup> )	1620	1750	1470	1110

B- 4: Polystyrene (manufactured by Asahi Kasei Industrial Inc.. "Stylon" 683)

[0048]

[Effect of the Invention]

The polyphenylene sulfide resin composition of this invention is the PPS resin excellent in the tracking resistance, without damaging excellent heat resistance, electric insulation and low water absorption properties which the PPS resin originally owns.